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# STRATOSPHERIC AEROSOL MODIFICATION BY SUPERSONIC TRANSPORT OPERATIONS WITH CLIMATE IMPLICATIONS

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## SUMMARY

The potential effects on stratospheric aerosols of supersonic transport emissions of sulfur dioxide gas and submicron-size soot granules are estimated. Recently, exhaust particles from large aircraft have been characterized experimentally; these new data have been adopted where appropriate. An interactive particle-gas model of the stratospheric aerosol layer is used to calculate changes due to exhaust emissions, and an accurate radiation transport model is used to compute the effect of aerosol changes on the Earth's average surface temperature. It is concluded that the release of large numbers of small soot particles into the stratosphere should *not* lead to a correspondingly significant increase in the concentration of large, optically active aerosols. On the contrary, the increase in large particles is severely limited by the total mass of sulfate available to make large particles in situ and by the rapid loss of small seed particles via coagulation. It is shown that a fleet of several hundred supersonic aircraft, operating daily at 20 km, could produce about a 20% increase in the concentration of large particles in the stratosphere. Moreover, aerosol increases of this magnitude would reduce the global surface temperature by less than 0.01 K; the climatic implications of a temperature change of this magnitude are negligible.

## 1. INTRODUCTION

There is increasing interest in the role of terrestrial aerosols in the radiation budget and climate of Earth, and in the possibility that civilization may be disturbing the natural balance of airborne particulates (ref. 1). For this reason, the effects on stratospheric aerosols of aircraft emissions of sulfur gases and soot particles were considered during the Climatic Impact Assessment Program (ref. 2). It was estimated that a fleet of advanced supersonic transports (SSTs) emitting $3\times10^7$ kg of $SO_2$ per year between 18 and 21 km might reduce the Earth's surface temperature by about 0.05 K. However, the CIAP conclusions were based on a simple residence time model for an aerosol of fixed-size dispersion; accordingly, they were somewhat uncertain.

Pollack et al. (refs. 3, 4) also investigated the climatic effect of sulfur dioxide gas released by SSTs. They found that the injection of $3\times10^7$ kg of $SO_2$ per year between 18 and 21 km might decrease the average global surface temperature by about 0.006 K. For several reasons, this temperature change is much smaller than that estimated by CIAP (ref. 2). The ambient aerosol layer assumed by Pollack et al. consisted of smaller (more transparent) particles on the average, and the

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related temperature calculations accounted for infrared radiation trapping by aerosols, which was neglected by CIAP. In addition, Pollack et al. omitted certain feedback mechanisms, such as increased ice formation leading to enhanced surface albedo, from their model. Finally, Pollack et al. adopted a residence time for particles in the altitude range from 18 to 21 km that was about one-half of the CIAP value. The accuracy of the aircraft calculations made by Pollack et al. were, however, limited by an incomplete treatment of aerosol physics and the neglect of soot emissions.

In the present study, these earlier investigations are expanded. Specifically, we consider the effect on the stratospheric aerosol layer of exhaust emissions from a hypothetical fleet of advanced SSTs. For calculating aerosol changes, a new and detailed model of the aerosol layer (refs. 5, 6) is used and, for determining the related effect on global surface temperatures, an improved radiation balance model (refs. 3, 4, 7) is used. We have chosen to study the stratospheric aerosol layer because it is believed to be quite sensitive to small anthropogenic perturbations (refs. 6, 8, 9) and is thought to influence the global climate (refs. 4, 7). Moreover, the ambient characteristics of the layer are relatively well established observationally (refs. 10–16).

The authors thank J. P. Friend of Drexel University, C. C. Chou of the University of California, Riverside, and L. E. Michalec of the Naval Air Rework Facility, North Island, San Diego, California, for use of their unpublished data.

## 2. SST ENGINE EMISSIONS OF GASES AND PARTICLES

The engines of SSTs emit gaseous sulfur oxides and carbonaceous particulates as exhaust constituents. The sulfur oxides are generated thermochemically from residual organic sulfur compounds found in most aviation fuels; they add indirectly to the natural aerosol loading of the atmosphere through condensation on preexisting particles. Soot, which is produced at high temperatures in the engine combustors, adds directly to atmospheric aerosol loading.

For the gaseous sulfur component, we adopt an emission index of 1 g of $SO_2$ per kilogram of fuel from CIAP (ref. 2). This is only a rough value based on an average amount of residual sulfur found in a wide variety of fuels. Inasmuch as sulfur is a recoverable component of such fuels, sulfur emissions to the stratosphere are readily controllable, although removal adds to the cost of the fuel. For our calculations, we assume an SST fleet of 300 aircraft of advanced engineering design. It has been estimated that each advanced SST would consume about 38,000 kg of fuel per hour and would be in operation 7 hr/day (e.g., see ref. 17). Accordingly, the assumed fleet of 300 aircraft would release about $2.9 \times 10^7$ kg of $SO_2$ per year worldwide.

The $SO_2$ emissions are averaged over the globe because advanced SSTs are expected to fly in both hemispheres; the $SO_2$ is injected at 20 km, a likely cruise altitude for future supersonic aircraft (ref. 17). Horizontal averaging of exhaust emissions is necessary because the analysis is made with a one-dimensional model. Normally, the zonal dispersion of injected pollutants occurs within a few days, while meridional spreading may take a few months. By comparison, stratospheric residence times for gases and aerosols are a year or more (ref. 2). Hence, global-scale averaging of continuously or frequently injected materials is a reasonable assumption in a study of global responses.

The soot emission of a J79-GE-10B jet engine, burning 227 kg of fuel at military power in a sea-level test chamber has been measured by the Naval Environmental Protection Support Service (ref. 18); the observed size distribution is shown in figure 1. Our extrapolation of the measured particle abundances to smaller sizes is also shown in the figure. The corresponding emission index, about 0.3 g soot/kg fuel, is larger by a factor of 3 than that previously estimated for advanced aircraft by Grobman and Ingebo (ref. 19).

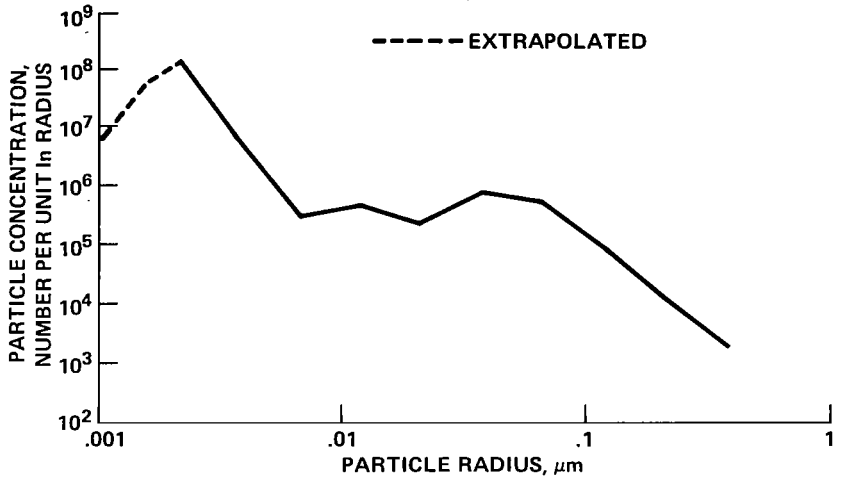


Figure 1.– Differential size distributions of soot particles in 22,700 $m^3$ of jet engine exhaust.

The rate of mass addition to the aerosol layer due to soot emission, about 0.3 g per kilogram of fuel, is much less than that due to sulfur emission, about 2 g per kilogram of fuel once the $SO_2$ is converted into a sulfuric acid-water solution with a 75% $H_2SO_4$ weight fraction (corresponding roughly to the observed composition of stratospheric aerosols; see ref. 20). Despite their small mass, it is conceivable that many of the injected soot particles could serve as condensation nuclei and eventually grow into large aerosol droplets. Using model calculations, however, we later demonstrate that soot coagulates rapidly enough with existing particles to restrict its role as seed for new aerosols.

## 3. THE AEROSOL MODEL

A one-dimensional (1-D) model of gaseous and particulate atmospheric constituents is used to calculate the effects of SST effluents on the stratospheric aerosol layer. The numerical techniques used in the model are discussed in detail in reference 5. Briefly, the particles are cataloged into "bins" according to their radii and altitudes. The model has 35 radius categories ranging from 0.001 $\mu$m to 2.56 $\mu$m, with particle volume doubling between sizes; there are 30 altitude levels from 0 to 58 km at 2-km intervals. The number of particles in each size-altitude bin is evaluated at each time step during a calculation. Two types of particles are distinguished: aerosol droplets and condensation nuclei. The sizes of inclusions (cores) within the aerosol droplets are also computed; in the present model, these cores consist of condensation nuclei that have been nucleated into droplets, or that have coagulated with existing droplets. Cores can accumulate and coalesce within droplets; they are released when the droplets evaporate.

Table 1 gives the photochemical interactions of sulfur-bearing gases that are included in the model. The dominant source of sulfur for the (model) aerosol layer is OCS, which diffuses upward from the troposphere to the stratosphere where it is photolyzed into S and CO (ref. 38); the existence of OCS in the stratosphere has recently been confirmed by the measurements of E. C. Y. Inn and coworkers (ref. 39). The subsequent rate of formation of $H_2SO_4$ vapor, which is a precursor of sulfate aerosol particles, is limited principally by the reaction of $SO_2$ with OH (R7 in table 1). The reaction scheme in table 1, although simple, reproduces many of the presently accepted general features of stratospheric sulfur photochemistry; for more information, see references 5 and 40.

TABLE 1.– REACTIONS OF SULFUR COMPOUNDS

| | Reaction | Rate coefficient[a] | Reference |
|---|---|---|---|
| R1 | $S + O_2 \rightarrow SO + O$ | $2.2\times10^{-12}$ | 21 |
| R2 | $SO + O_2 \rightarrow SO_2 + O$ | $3.0\times10^{-13}\ e^{-2800/T}$ | 22 |
| R3 | $SO + O_3 \rightarrow SO_2 + O_2$ | $2.5\times10^{-12}\ e^{-1050/T}$ | 22 |
| R4 | $SO + NO_2 \rightarrow SO_2 + NO$ | $1.5\times10^{-11}$ | 22 |
| R5 | $SO_2 + O + M \rightarrow SO_3 + M$ | $3.4\times10^{-32}\ e^{-1130/T}$ (M=$N_2$) | 23[b] |
| R6 | $SO_2 + HO_2 \rightarrow SO_3 + OH$ | $1.0\times10^{-13}\ e^{-1410/T}$ | 26[c] |
| R7 | $SO_2 + OH + M \rightarrow HSO_3 + M$ | $8.2\times10^{-13}/(7.0\times10^{17} + [M])$ (M=$N_2$, 220 K) | 27[d] |
| R8 | $HSO_3 + OH \rightarrow SO_3 + H_2O$ | $1\times10^{-11}$ | (e) |
| R9 | $SO_3 + H_2O \rightarrow H_2SO_4$ | $9.1\times10^{-13}$ | 28 |
| R10 | $OCS + O \rightarrow SO + CO$ | $3.0\times10^{-11}\ e^{-2270/T}$ | 29[f] |
| R11 | $SO_2 \rightarrow$ washout | $3.8\times10^{-6}\ \frac{(13 - z)}{13}$ $z \leqslant 13$ km<br>0 $z > 13$ km | (g) |
| R12 | $HSO_3 \rightarrow$ washout | $R_{12} = R_{13}$ | (h) |
| R13 | $H_2SO_4 \rightarrow$ washout | $2.3\times10^{-5}\ \frac{(13 - z)}{13}$ $z \leqslant 13$ km<br>0 $z > 13$ km | (i) |
| $J_1$ | $SO_2 + h\nu \rightarrow SO + O$ | $1.9(-13)^j$, 3.1(-10), 2.3(-8), 4.1(-7), 2.7(-6), 8.5(-6), 1.8(-5), 2.6(-5), 3.3(-5) | 34<br>35 |
| $J_2$ | $OCS + h\nu \rightarrow S + CO$ | $1.5(-14)^j$, 1.9(-11), 1.4(-9), 3.0(-8), 2.8(-7), 1.4(-6), 5.0(-6), 1.1(-5), 1.7(-5) | 36[k] |
| $J_3$ | $H_2SO_4 + h\nu \rightarrow SO_2$ + products | $8.5(-16)^j$, 1.6(-12), 1.3(-10), 2.4(-9), 1.5(-8), 4.4(-8), 8.6(-8), 1.3(-7), 1.6(-7) | (l) |

[a]Rate coefficient units are $sec^{-1}$ for unimolecular, $cm^3\ sec^{-1}$ for bimolecular, and $cm^6\ sec^{-1}$ for termolecular processes.

[b]Atkinson and Pitts (ref. 24) have obtained a similar rate constant, $9.2\times10^{-32}\ e^{-(1000\pm200)/T}\ cm^6\ sec^{-1}$, for M = $N_2O$, and Westenberg and deHaas (ref. 25), a value of $1.1\times10^{-31}\ e^{-(1400\pm50)/T}\ cm^6\ sec^{-1}$ for M = He.

[c]The rate constant measured at room temperature is $9\times10^{-16}\ cm^3\ sec^{-1}$; we have assumed a prefactor of $1\times10^{-13}\ cm^3\ sec^{-1}$ and the corresponding activation energy. Reaction R6 is much less important than reaction R7 in the aerosol layer.

[d]The rate constant value is based on a review of the available measurements, principally those by Castleman et al. (ref. 28). The observed pressure dependence has been simulated by a two-state reaction mechanism.

[e]An estimated value; complemented by R9, reaction R8 is proposed to replace a complex, but as yet undetermined, photo-reaction sequence starting with $HSO_3$ and presumably leading to sulfuric acid vapor production.

[f]Krezenski et al. (ref. 30) have obtained a similar rate coefficient of $1.6\times10^{-11}\ e^{-2250/T}\ cm^3\ sec^{-1}$.

[g]The assumed $SO_2$ washout rate is based on an estimated low-altitude residence time of about 3 days (refs. 31–33).

[h]Equal washout rates for $HSO_3$ and $H_2SO_4$ are assumed.

[i]The $H_2SO_4$ washout rate is based on an assumed 0.5-day residence time near the surface.

[j]Twenty-four-hour average photodissociation rates ($sec^{-1}$) are given for altitudes of 10, 15, 20, 25, 30, 35, 40, 45, and 50 km, respectively. The number 1.9(-13) $\equiv 1.9\times10^{-13}$.

[k]OCS cross sections at 232 K were also obtained from C. C. Chou, University of California, Riverside, Calif. (private communication, 1976).

[l]The $H_2SO_4$ photoabsorption spectrum is assumed to be the same as the HCℓ spectrum with cross-section data taken from reference 37. Ultraviolet decomposition of $H_2SO_4$ in the upper stratosphere is assumed to lead to $SO_2$ production.

Some new laboratory evidence suggests that in the atmosphere sulfur radicals, such as $HSO_3 \cdot O_2 \cdot H_2O$ (or $H_3SO_6$), will be formed following the reaction of $SO_2$ with OH (R7, table 1). Moreover, clusters of only two of these radicals can apparently act as nuclei for water vapor condensation in a supersaturated "cloud" chamber. (Private communication from J. P. Friend, Drexel University.) The sulfur radicals, upon entering a solution droplet, or possibly even in the molecular phase, should react rapidly to form $H_2SO_4$ and other sulfates. We are led to this conclusion by the fact that $SO_4$ is found to be the dominant oxidation state of sulfur in collected aerosol samples, and $H_2SO_4$-$H_2O$ solutions have most of the observed spectroscopic and physical properties of natural aerosols (refs. 11, 12, 15, 20). After studying the changes in our model predictions caused by allowing sulfur radicals ($HSO_3$) to condense directly onto aerosols, we conclude that as long as $H_2SO_4$ is the end product of sulfur radical chemistry, our approach assuming $H_2SO_4$ as the precursor of aerosol droplet formation and growth is appropriate for calculating the properties of aerosols with radii larger than 0.01 $\mu$m. In order to be able to predict the concentrations of sulfur radicals in air as well as their precise effects on aerosol size dispersion and composition, however, we will have to incorporate species such as $H_3SO_6$ in our model, when additional information about their photochemistry and solution properties becomes available.

Suitable theories are available to describe the nucleation, growth, and coagulation of aqueous sulfuric acid solution droplets under stratospheric conditions. In our model, nucleation of supersaturated $H_2SO_4$ vapor occurs on the surfaces of condensation nuclei (refs. 41, 42); the condensation nuclei are transported by eddy diffusion from the troposphere to the stratosphere. Whenever

these nuclei are mixed into a supersaturated stratospheric environment, or are deposited in such a region by aircraft engines, they are assumed to be nucleated in $10^6$ sec. We are currently evaluating the influence on natural aerosol formation of $H_2SO_4$ homogeneous nucleation and nucleation onto ions, and of sulfur radical nucleation; preliminary results suggest that these processes are more critical in the troposphere than in the stratosphere.

Aerosol droplets can grow by heteromolecular condensation of sulfuric acid and water vapors. A growth equation is used here that is based on the formulation of molecular diffusion to particles developed by Fuchs and Sutugin (ref. 43), suitably modified for heteromolecular condensation (ref. 42). Coagulation is a growth mechanism that for small droplets can be more important than condensation. Accordingly, we have developed a detailed coagulation algorithm for our model, utilizing the coagulation kernels derived by Fuchs (ref. 44). Particle gravitational sedimentation rates are calculated using the Stokes-Cunningham equation for the terminal velocity of spherical droplets in air (ref. 45). Vertical particle diffusion rates are computed with an "eddy" diffusion coefficient which is illustrated in reference 5.

The lifetime of an aerosol droplet against washout in the troposphere is assumed to be 1 day near the ground, with the inverse lifetime decreasing linearly to zero at the tropopause level (situated near 13 km in our model). For aerosol droplets in the upper troposphere, this leads to an effective washout lifetime of about 1 week.

Most of the physical and chemical processes that are treated in our aerosol model are summarized schematically in figure 2.

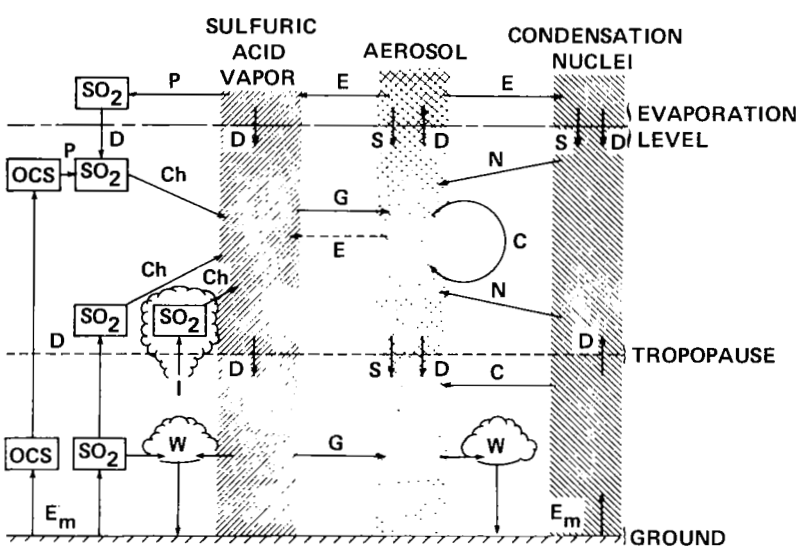


Figure 2.– Schematic outline of physical and chemical interactions included in one-dimensional model.

## 4. AEROSOL MODEL PREDICTIONS: THE AMBIENT AND PERTURBED STRATOSPHERE

Model simulations of the ambient stratospheric aerosol layer, and detailed tests of model sensitivity to a large number of physical parameters, are presented by Toon et al. (ref. 6). The model predictions are in good agreement with observational data for the following aerosol parameters: the height distribution and magnitude of the total particle mass mixing ratio; the vertical concentration profile of total particulates; the aerosol size distribution; the mixing ratio of large aerosol particles (radii $> 0.15$ $\mu$m); the number ratio of particles with radii $> 0.15$ $\mu$m to those with radii $> 0.25$ $\mu$m; and the aerosol-droplet, sulfuric-acid weight fraction.

In figure 3, for example, we compare calculated and observed large-particle aerosol mixing ratios in the stratosphere. The measurements shown in figure 3 were made at northern midlatitudes during 1972-1973 with balloon-borne optical particle counters that discriminate against particles with radii $< 0.15$ $\mu$m (refs. 13, 14). Shown in figure 3 are the mean value of the data and the edge of the envelope containing the individual measurements. Individual measurements show that the layer is highly stratified with 50% variations in mixing ratios over 1-km vertical distances. The stratospheric data indicate little seasonal trend. A mixing ratio maximum generally occurs at about 24 km at the equator and at 17 km over the poles. The maximum mixing ratio is 50% greater at the equator than at the poles. The data from 1972-1973 were used because the total number of

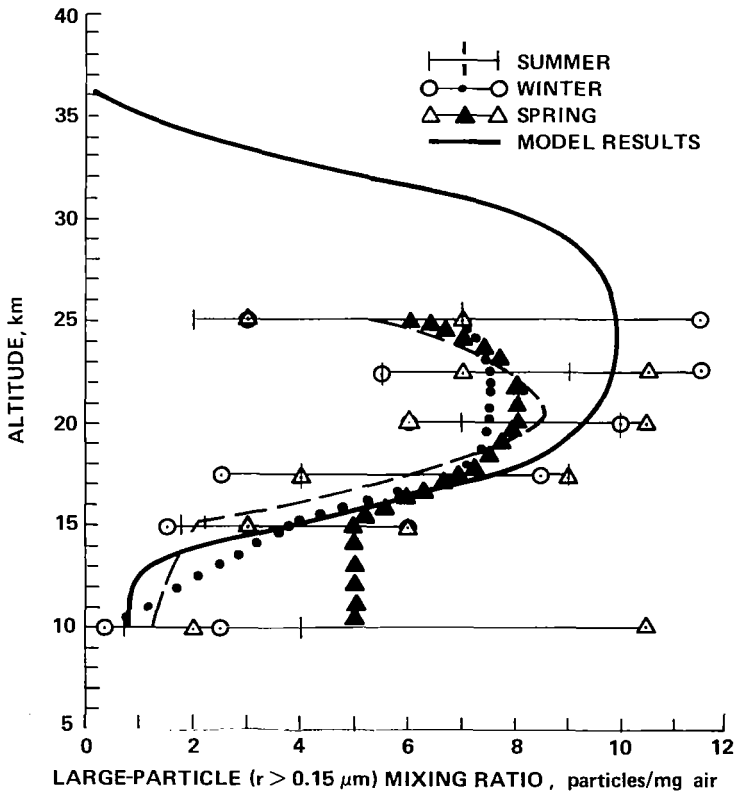


Figure 3.– Comparison of ambient model prediction with observations for large-particle number mixing ratio (i.e., number of particles per milligram of air with radii $> 0.15$ $\mu$m).

particles was near a minimum and the data were thus the least volcanically affected data that were available.

As figure 3 shows, the model prediction appears to represent a reasonable average of the observational values between 15 and 25 km (see ref. 6 for a complete discussion of model comparisons with data). Incidentally, it is the large particles that dominate the radiation transport properties of aerosols.

The effect of SST emissions on the large-particle mixing ratio is shown in figure 4. The figure shows calculated steady-state mixing ratios for large particles (radii $> 0.15$ $\mu$m) in the stratosphere for SST flights both with and without soot emission; the assumed cruise altitude is 20 km. The global exhaust injection rate for $SO_x$ is $2.9 \times 10^7$ kg of $SO_2$ per year; the rate for soot is $8.7 \times 10^6$ kg of soot per year. The size distribution of the soot particles is as shown in figure 1. For purposes of comparison, the ambient model large-particle mixing ratio profile and some observational data are also given in figure 4. For each profile, the total stratospheric column concentration of large particles is shown in parentheses. (Note that all of the perturbation calculations reported here correspond to 5 years of continuous injection of contaminants; for aerosols, these are essentially steady-state calculations because we verified that extension of simulation times to 10 years causes less than a 10% adjustment in the calculated perturbations.)

The most pronounced effect of SSTs is due to sulfur gas emission, not soot emission. The soot particles, which are quite small on the average, coagulate rapidly with aerosols of all sizes; likewise,

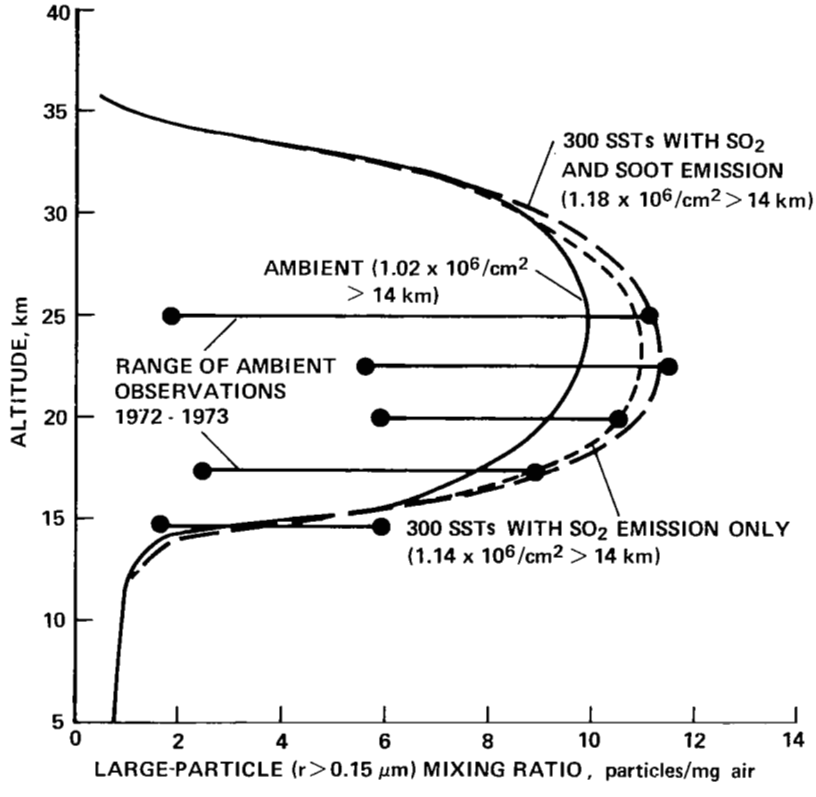


Figure 4.– Supersonic transport effects on large aerosol particles (radii $> 0.15$ $\mu$m).

sulfur gas is absorbed by existing particles of all sizes. Therefore, the overall effects of soot and $SO_2$ on existing aerosols are alike in that both pollutants cause the existing particles to grow in size, with their relative importance being largely determined by the relative mass injected. Interestingly, the predicted change in aerosol concentration lies within the range of variability (or uncertainty) in current measurements (see fig. 4), and might therefore be difficult to detect. Also note in figure 4 that SST emissions affect the entire aerosol layer because of vertical dispersion by diffusion from the height of injection (20 km in this case).

The addition of very small soot particles to the stratosphere could, in theory, increase the abundance of aerosols with radii of 0.1 to 0.3 $\mu$m, despite the small total mass added. This is so because small particles with large effective surface areas efficiently absorb injected sulfur gas as they grow and coagulate into the middle size range (radii $\sim$ 0.1–0.3 $\mu$m). The number of large particles produced by soot emission would be maximized if coagulation did not occur and if the sulfur gas supply were sufficient to allow every newly formed droplet to grow at a rate as fast as 0.5 $\mu$m per year. However, the coagulation lifetime ($\sim$1 month) of added soot particles and the small aerosols that are nucleated on them is much shorter than the time required to grow a large particle ($\sim$1 year). Moreover, as Toon et al. (ref. 6) have pointed out, the natural supply rate of sulfur to the stratosphere via transport from the lower atmosphere is quite restricted, as is the supply rate of injected sulfur for the pollution cases treated here; this also limits the number of new particles that can grow to large sizes in the model. It is unlikely that the natural supply rate of sulfur to the aerosol layer is much larger than that simulated by our model (equivalent to about $1\times10^5$ to $2\times10^5$ metric tons of $SO_2$ per year) because of the implications for the observable properties of the ambient aerosols. Accordingly, the release of soot particles in the stratosphere can only result in a small increase in large particles, by the mechanism of combined growth and coagulation described above.

Novakov et al. (ref. 46) have demonstrated that soot particles can efficiently oxidize $SO_2$ into sulfate via a catalytic process involving oxygen molecules adsorbed on the particle surfaces. Soot particles released in the stratosphere could therefore act as centers for heterogeneous nucleation and heterogeneous chemical formation of sulfates. Nevertheless, the end result is still the same, as these particles (whether they are dry soot or soot particles coated with sulfuric acid) will quickly coagulate with each other and with preexisting aerosol particles. Because aerosol production is limited by the availability of sulfur-bearing gases in the stratosphere, the total aerosol mass is largely independent of the nucleation mechanisms operating. Further, as implied earlier, an increased rate of $SO_2$-to-sulfate conversion would not greatly affect the model predictions made here (precluding the possibility of rapid particle formation in the near-wake of the aircraft). In the upper troposphere, on the other hand, where larger quantities of soot are continually supplied by aircraft traffic, and where $SO_2$ is much more abundant, a substantial number of new aerosol particles might be formed around soot. This problem is beyond the scope of the present work, however.

With regard to SSTs, then, sulfur gas emissions cause a much larger increase in the total aerosol mass and large-particle concentration than soot emissions. It should be noted, by comparing the results in figure 4, that if the sulfur component of aviation fuel were eliminated, the net effect of soot emission alone on large aerosol particles would be quite small (the soot effect is roughly independent of $SO_2$ emission for the injection levels considered).

Further insight into the effects of SSTs on particles in the aerosol layer can be gained from figures 5 and 6. Figure 5 shows that the total number of stratospheric particles is increased only slightly by SSTs, despite the large number of small particles injected. In figure 6, the total number of soot particles of each size that is injected into 1 $cm^3$ of air at 20 km during 1 year of flight activity is compared with the ambient and perturbed steady-state particle size distributions. The comparison illustrates that because the soot particles are injected near 20-km altitude, where fairly large numbers of acid droplets exist, the soot is rapidly depleted by coagulation. Note in figures 1 and 6 that the soot size distribution is somewhat deficient in the particle-size range between 0.01 and 0.03 $\mu$m. Particles in this range would be quite efficient (per unit mass injected) in producing additional large stratospheric particles. The increase in aerosol mass caused by SST emissions is difficult to discern in figure 6 because it is only ~20% (similar to the increase in large particles shown in fig. 4), which implies only ~5% increase in the average particle radius.

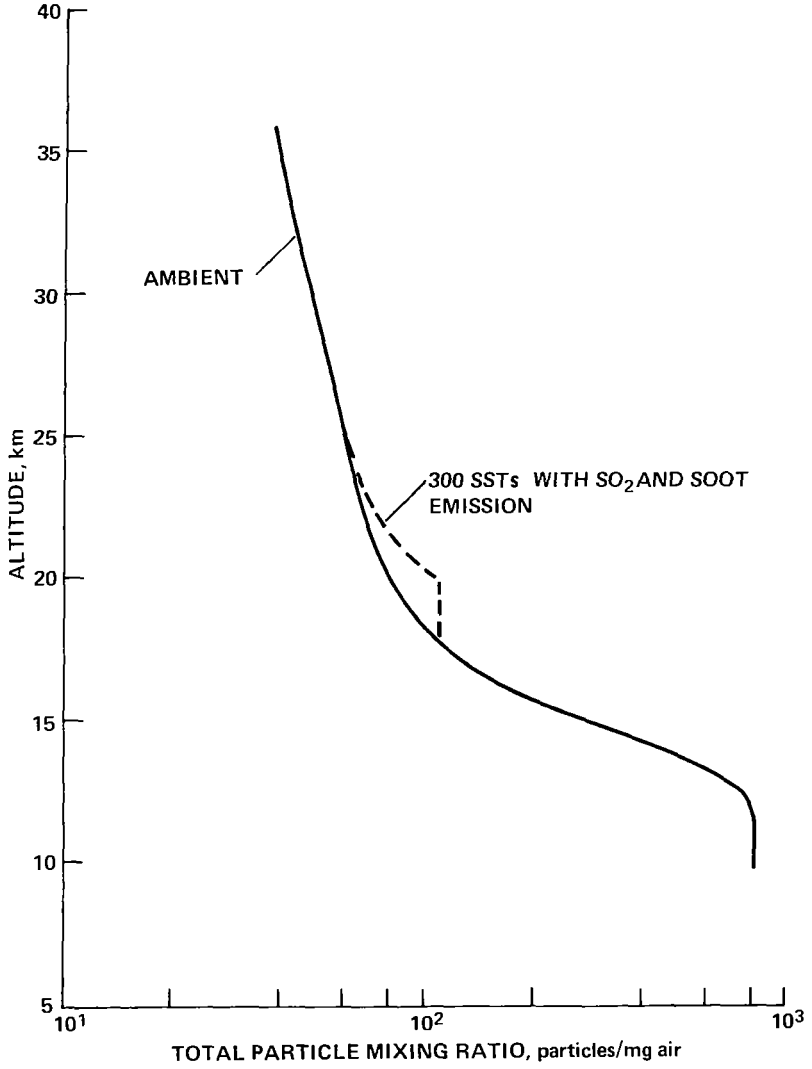


Figure 5.— Supersonic transport effects on total mixing ratio of stratospheric particles.

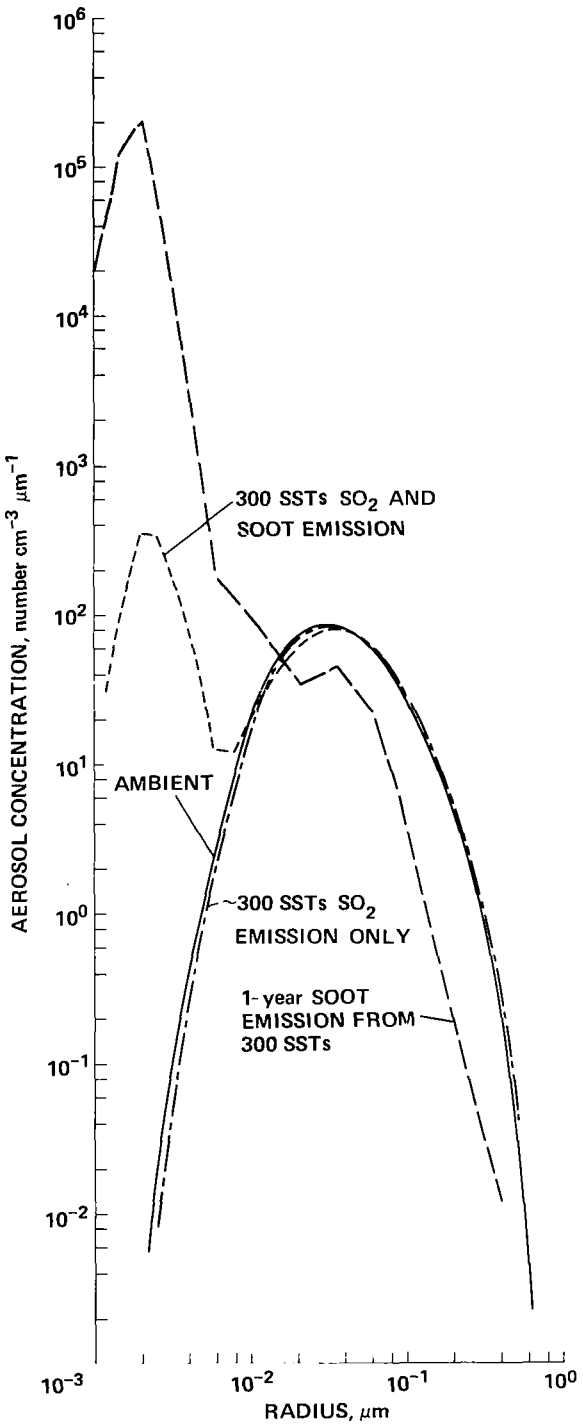


Figure 6.– Predicted aerosol particle size distribution at 20 km for SST injection cases shown in figure 4.

## 5. RADIATIVE TRANSFER CALCULATIONS OF CLIMATE EFFECTS DUE TO AEROSOL PERTURBATIONS

In previous studies, Pollack et al. (refs. 3, 4) considered the climatic effect of $SO_2$ injected by various numbers of SSTs flying in the stratosphere. The present study represents an improvement over this earlier work in several respects.

First, Pollack et al. (refs. 3, 4) were not able to model the stratospheric aerosols. Therefore, they could not consider the interaction of soot emissions with ambient stratospheric aerosols. Moreover, they had to assume a fixed stratospheric particle size distribution, and could only roughly estimate optical depth changes due to exhaust emissions because of particle residence time variations with altitude. Here we include both the gaseous and particulate emissions of SSTs, and explicitly treat gas-particle and particle-particle interactions. The present model, in addition, predicts most of the physical parameters needed to make an accurate calculation of aerosol optical properties.

The second major improvement in this work is that a fully convergent radiative-convective solution for the vertical temperature profile is used. In our earlier work we assumed that the shape of the temperature profile was invariant, and calculated the change in surface temperature by achieving a flux balance between incoming solar energy and emitted infrared radiation at the top of the atmosphere.

To calculate the climatic effects of aerosols injected by SSTs, the altitude-dependent size distributions predicted by our aerosol model is used in the doubling routine described by Pollack et al. (ref. 3). The doubling calculations are highly accurate multiple scattering computations that explicitly account for solar energy absorption by $CO_2$, $O_3$, $O_2$, and $H_2O$, and absorption and scattering by aerosols in the stratosphere and troposphere. In this way, the altitude-dependent solar energy deposition rate for perturbed and

unperturbed conditions is obtained. For stratospheric aerosols the optical constants of a 75% sulfuric acid aqueous solution are used, and for condensation nuclei the optical constants of $(NH_4)_2SO_4$ are used. Of course, the soot grains from SSTs have optical constants that differ from natural condensation nuclei. Most atmospheric condensation nuclei are found, however, in the troposphere; the fraction residing in the stratosphere, either by number or mass, is extremely small, both for ambient and perturbed conditions. Accordingly, our results are not sensitive to the optical constants assumed for soot. For tropospheric particles, which are held fixed, the model of Toon and Pollack (ref. 15) is used.

Once the profile of the solar energy deposition rate is determined from the doubling calculations, an infrared radiation transport calculation is performed to achieve radiative-convective equilibrium. This routine uses the same numerical techniques described by Pollack et al. (ref. 3) to calculate the infrared radiation emitted and absorbed by $H_2O$, $CO_2$, $O_3$, and aerosols. Scattering by aerosols is ignored because of the small single-scattering albedo. The routine iterates to obtain radiative equilibrium in the stratosphere with relative humidity, cloudiness (50% cloud cover), and the top height of clouds held fixed. Convergence is assumed when the difference between the net upward infrared heat flux and the total solar flux absorbed below a given level is less than $10^{-6}$ of the solar flux absorbed below that level. The surface temperature change between the next to last and the final iteration is always much less than $10^{-3}$ K, although at high altitudes (above 50 km) temperature changes are sometimes several tenths of a degree; the slower temperature convergence above 50 km has a negligible effect on convergence at the ground.

When compared with existing atmospheric conditions, we find that a fleet of 300 SSTs flying 7 hr/day at 20 km would be expected to cause a global surface temperature decrease of the order of $3\times10^{-3}$ K. (Between the ambient and perturbed solutions, at the final iteration no differences in computed temperatures larger than $10^{-2}$ K were found at higher altitudes, and even those might have been reduced if the convergence had been carried out further.) By comparison, the data of Pollack et al. (refs. 3, 4) suggest that the same number of aircraft could lower the surface temperature by about $6\times10^{-3}$ K. Evidently, our more sophisticated calculations confirm that the earlier estimates of Pollack et al. were quite reasonable.

## 6. UNCERTAINTIES IN THE PREDICTIONS

From the preceding discussion, it should be apparent that there are numerous sources of uncertainty in our calculations. Due to a lack of detailed information, it is difficult to quantify such uncertainties in a statistical sense. Accordingly, we will only outline the major uncertainties and make some subjective estimates of their magnitudes to guide the reader. It should be kept in mind that in many areas of atmospheric science calculations bearing uncertainty factors as large as 2 or 3 are considered to be quite accurate.

Aerospace engine emissions are not very well defined (see our previous discussions of the data). For SSTs, the rate of soot ejection is uncertain by a factor of at least 2. Moreover, the number of small soot particles is undetermined to within a factor of 2 or 3. The $SO_2$ emission index varies widely between aviation fuel lots and could differ by a factor of 2 from our adopted value of 1 g of $SO_2$ per kilogram of fuel. Careful measurements of these exhaust parameters using prototype aircraft engines would help to reduce some of the related assessment uncertainties.

The aerosol model introduces additional uncertainty into the calculations; most of these uncertainty sources are discussed by Turco et al. (ref. 5) and Toon et al. (ref. 6). Because the model has a one-dimensional structure and incorporates physical parameters corresponding to middle latitudes, it presumably represents average global or hemispherical conditions. Although this interpretation of the model is only partially valid, Turco et al. and Toon et al. argue that the global uniformity of the stratospheric aerosol layer allows an accurate description with a one-dimensional model. Even so, many of the components of normal temporal and geographical variability cannot be treated with such a model; as a result, only globally averaged steady-state predictions are given here. There is, in addition, the question of the significance of in situ aerosol nucleation from injected sulfur gases, with which we have not yet dealt exhaustively; however, it seems unimportant based on our simulations of large stratospheric injections of soot particles (which act as condensation nuclei). Despite these reservations, however, we feel that the aerosol model reasonably simulates the natural sources and sinks of stratospheric sulfate particles, which is a critical factor in this assessment (see the discussion in sec. 4). Our computed aerosol changes are, therefore, probably uncertain by a factor of no more than 3.

Uncertainties in the radiation transport calculations are carefully outlined in the papers by Pollack et al. (refs. 3, 4, 7). The current improved radiation calculations are thought to be accurate to within a factor of 2.

We estimate an overall uncertainty of a factor of 5 for our predicted global temperature reductions caused by aerospace activity, although unidentified sources of systematic uncertainty outside this range are not precluded. Although the overall uncertainty is large, it is not unusual in atmospheric science. Inasmuch as the predicted climate changes are far below the threshold of detection, large uncertainties in the magnitude of the changes should have little significance.

## 7. CONCLUSIONS

We have made some new, and more detailed, calculations of the effects of supersonic transport exhaust emissions on the stratospheric aerosol layer and on the surface temperature of Earth. For this purpose, an interactive gas-particle model of the sulfate aerosol layer and a comprehensive simulation of terrestrial radiation transport processes were used. Gaseous and particulate emissions of aircraft engines, both of which might affect the natural aerosols, were considered. Basically, we conclude that, although likely future levels of aerospace activity could exert a moderate influence on the stratospheric aerosol layer, such activity would have a negligible effect on the climate of Earth. For example, 300 SSTs used in daily service are estimated to increase the large particle concentration of the stratosphere by only about 20% and to decrease the planet's average surface temperature by less than 0.01 K. These estimates are based on computer model simulations and must, accordingly, be considered as tentative. Interestingly, despite the greater sophistication of the models used here, our new predictions are very close to those made several years ago by Pollack and coworkers (refs. 3, 4).

With the more complete models used in this work, however, it has been possible to resolve several issues related to the effects of large-scale injections of very fine particles into the stratosphere. It was found that the number of small particles that can grow freely to large size in the stratosphere is quite limited. Indeed, the total number of large particles is restricted by the supply

rate of sulfur to the aerosol layer. This rate, presently controlled by natural processes, amounts to about $1\times10^5$ to $2\times10^5$ metric tons of $SO_2$ per year (refs. 11, 13); by contrast, only about $0.3\times10^5$ metric tons of $SO_2$ per year would be released by several hundred advanced SSTs. Because of the dominance of natural processes, the total mass of the sulfate layer and the number of large particles comprising it are relatively invariant for the conditions studied. Most small soot particles added to the aerosol layer coagulate with larger particles. Thus, it appears that SST emissions of $SO_2$, which can be regulated if necessary, would have the greatest effect on stratospheric aerosols.

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15. *Supplementary Notes*

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16. Abstract

The potential effects on stratospheric aerosols of supersonic transport emissions of sulfur dioxide gas and submicron-size soot granules are estimated. An interactive particle-gas model of the stratospheric aerosol is used to compute particle changes due to exhaust emissions, and an accurate radiation transport model is used to compute the attendant surface temperature changes. It is shown that a fleet of several hundred supersonic aircraft, operating daily at 20 km, could produce about a 20% increase in the concentration of large particles in the stratosphere. Aerosol increases of this magnitude would reduce the global surface temperature by less than 0.01 K.

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